

Sum rule for a Schiff-like dipole moment

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The energy-weighted sum rule for an electric dipole transition operator of a Schiff type differs from the Thomas-Reiche-Kuhn sum rule by several corrective terms which depend on the number of system components, \mathcal{N} . The deviations are evaluated by two distinct approaches. One of them is based on the charge density expansion around an uniform distribution and provides an analytical \mathcal{N} dependence for the sum rule while, the other one hinges of the RPA approach and yields compact expressions for corrections as function of the RPA amplitudes. Although the formalism might be considered for any many body system, we applied it for illustration, to the case of Na clusters. One concludes that the RPA results for Na clusters obey the modified TRK sum rule.

The Thomas-Reiche-Kuhn (TRK) sum rule [1, 2, 3] has been widely used in various contexts of electron excitations in atoms, molecules, and solids. Indeed, the relation to the photoabsorbtion oscillator strength [4, 5, 6, 7] makes it quite useful in interpreting various collective features of the dipole states.

The many body properties like collective excitations in atoms, molecules or clusters are studied using different collective models. Thus, the random phase approximation (RPA) formalism has been extensively used and excellent results for the photoabsorbtion cross section spectra, especially in the atomic clusters domain [8, 9, 10] were obtained.

In a previous publication [10], some of the many body features of the small and medium sodium clusters were studied within the RPA approach using the projected spherical single-particle basis defined in Ref.[11]. The RPA wave functions were used to treat the dipole transitions which led to the photoabsorbtion cross section spectra. Also, the system static electric polarizability was analytically determined. The nice virtue of the RPA approach with spherical single particle basis consists of that it satisfies TRK sum rule.

This is however true for the electric dipole moment, which is not the case in the above quoted paper where, indeed, a modified dipole operator, similar with Schiff-like dipole moment [12, 13, 14, 15] is used. The corrections to the dipole operator accounts for the screening effect, caused by the electronic shells, on the motion of the valence electrons in the mean field determined by the nuclear charge. Moreover the approach of Ref.[10] uses a projected spherical single particle basis which allows for an unified description of spherical and deformed clusters. Here we address the question whether these specific features require a modification of the TRK sum rule.

Thus, in the present paper we derive a new sum rule which corresponds to the Schiff-like dipole moment.

Within the RPA formalism, for any Hermitian operator \hat{M} , the following sum rule holds

$$\sum_n (E_n - E_0) \left| \langle 0 | \hat{M} | 1_n \rangle \right|^2 = \frac{1}{2} \langle 0 | [[\hat{M}, H], \hat{M}] | 0 \rangle, \quad (1)$$

where E_n are the RPA energies associated to the many body Hamiltonian H . Here the state $|0\rangle$ is the RPA phonon vacuum, while $|1_n\rangle$ denotes the single phonon state $|1_n\rangle = C_n^\dagger |0\rangle$. We are interested in those features of atomic clusters which are determined by the valence electrons. These move in a mean field, determined by the ionic core, and interact among themselves through a Coulomb force. The residual two-body interaction [16, 17] can be expanded in multipole series from which only the dipole term is relevant and therefore considered. The many body Hamiltonian describing the system is treated within the RPA formalism [10], which defines the phonon operator:

$$C_n^\dagger(1, \mu) = \sum_{ph} \left[X_{ph}^n (c_p^\dagger c_h)_{1\mu} - Y_{ph}^n (c_h^\dagger c_p)_{1\mu} \right], \quad (2)$$

by means of the equations:

$$[H, C_n^\dagger] = E_n C_n^\dagger, \quad (3)$$

$$[C_n, C_{n'}^\dagger] = \delta_{nn'}. \quad (4)$$

Here X_{ph}^n and Y_{ph}^n are the RPA forward-going and backward-going amplitudes corresponding to the n -th order solution of the RPA equations. The reduced probability for the dipole transition $|0\rangle \rightarrow |1_n^+\rangle$ can be written in terms of the RPA phonon amplitudes and the ph matrix elements of the transition operator [1] :

$$B(E1, 0^+ \rightarrow 1_n^+) = \left| \langle 0 | \mathcal{M}(E1) | 1_n^+ \rangle \right|^2, \quad (5)$$

where

$$\begin{aligned} \langle 0 | \mathcal{M}(E1) | 1_n^+ \rangle &= \sum_{ph} \hat{I}_p \langle p | \mathcal{M}(E1) | h \rangle \\ &\quad [X_{ph}^n + (-)^{I_p + I_h} Y_{ph}^n] \end{aligned} \quad (6)$$

are the reduced matrix elements of the dipole operator $\mathcal{M}(E1)$, between the specified RPA states.

[1] Throughout this paper the Rose's convention for the reduced matrix elements are used.

Instead of the usual transition dipole operator, a Schiff-like moment operator [12, 13, 14, 15] was used.

$$\mathcal{M}(E1) = e \left(1 - \frac{3}{5} \frac{r^2}{r_s^2} \right) \vec{r}. \quad (7)$$

Here r_s is the Wigner-Seitz radius and have the value of 3.93 a.u. for Na clusters. The corrective component, involved in the dipole operator, relates particle and hole states characterized by $\Delta N = 3$, which results in modifying the strength distribution among the RPA states. Such an effect would be however obtained even for the dipole transition operator, if the mean field potential for the single particle motion involves higher powers of the radial coordinate.

Reckoning the double commutator from Eq. (1), corresponding to the transition operator (7) one obtains:

$$\sum_n (E_n - E_0) |\langle 0 | \mathcal{M}(E1) | 1_n \rangle|^2 = \frac{9\hbar^2 e^2}{2m_e} \times \left[\mathcal{N} - \frac{2}{r_s^2} \langle 0 | \sum_{\alpha=1}^{\mathcal{N}} r_{\alpha}^2 | 0 \rangle + \frac{33}{25r_s^4} \langle 0 | \sum_{\alpha=1}^{\mathcal{N}} r_{\alpha}^4 | 0 \rangle \right]. \quad (8)$$

The terms correcting the standard TRK sum rule are the expected values of the radius powers r^2 and r^4 , in the RPA ground state. Obviously, these corrective terms induce an additional \mathcal{N} dependence for the energy-weighted sum rule. As for the left hand side of Eq.(8), this can be directly calculated using the RPA output data, like energies and transition probabilities [10]. The terms from the right hand side of Eq.(8) are evaluated alternatively through two distinct methods, which are briefly presented below.

A) *The boson expansion method.* The correction terms from the right hand side of the equation (8), which will be hereafter denoted as $S(\mathcal{N})$, can be expressed in terms of particle-particle (pp) and hole-hole (hh) transition components. Indeed, the ph transition components give vanishing contributions when they are averaged with the RPA ground state. Therefore in the second quantization, the needed one-body operator \hat{r}^m can be written as:

$$\sum_{\alpha=1}^{\mathcal{N}} r_{\alpha}^m \equiv \sum_p \langle p | r^m | p \rangle c_p^{\dagger} c_p + \sum_h \langle h | r^m | h \rangle c_h^{\dagger} c_h. \quad (9)$$

Following the boson expansion procedure, the fermion density operators $c_p^{\dagger} c_p$ and $c_h^{\dagger} c_h$ can be expressed in terms of the RPA phonon operators $C_n^{\dagger} (C_n)$. Thus, the particle and hole density operators can be written as

$$c_p^{\dagger} c_p = \sum_n a_n^p C_n C_n^{\dagger}, \quad c_h^{\dagger} c_h = \sum_n b_n^h C_n C_n^{\dagger}, \quad (10)$$

where the coefficients a_n^p and b_n^h have the expressions:

$$a_n^p = \langle 0 | [[C_n^{\dagger}, c_p^{\dagger} c_p], C_n] | 0 \rangle, \quad b_n^h = \langle 0 | [[C_n^{\dagger}, c_h^{\dagger} c_h], C_n] | 0 \rangle. \quad (11)$$

In this way the modified dipole sum rule becomes:

$$\sum_n (E_n - E_0) |\langle 0 | \mathcal{M}(E1) | 1_n \rangle|^2 = \frac{9\hbar^2 e^2}{2m_e} [\mathcal{N} + \sum_{n,p,h} \left((X_{ph}^{(n)})^2 + (Y_{ph}^{(n)})^2 \right) \left[-\frac{2}{r_s^2} r^2(ph) + \frac{33}{25r_s^4} r^4(ph) \right]], \quad (12)$$

where the following notation has been used:

$$r^k(ph) = \langle p | r^k | p \rangle - \langle h | r^k | h \rangle, \quad k = 2, 4. \quad (13)$$

As we have already mentioned, a specific ingredient of the approach from Ref. [10] is the use of the projected spherical single particle basis in the RPA formalism.

B) *Electron density approach.* Static electric polarizabilities results based on the calculations for the number of spilled out electrons [10], agree quite well with the corresponding experimental data. These spilled out electrons produce a screening effect against external fields which results in changing the classical result for the polarizability. The basic assumption in accounting the spilled out electrons is the fact that the electron density is not going sharply to zero at the cluster surface, but is gradually decreasing and moreover extends significantly beyond the jellium edge. The same argument can be brought for the correction terms of the $S(\mathcal{N})$ containing averages of the radius powers with the RPA vacuum states. The electronic density has a constant central part, enfolded by a diffuse region, of a width equal to a , where the electron density tends smoothly to zero. Guided on some parallelism between atomic clusters and nuclear systems [18, 19], the average of r^m with the RPA vacuum state is approximated by folding r^m with a localization probability density with spherical symmetry, of Fermi distribution type, having a profile similar to that of the charge density, mentioned above. In this way the average may be written in the form of a power series in the variable $\frac{a}{R}$ [20]:

$$\langle r^m \rangle = R^m \frac{3}{m+3} \left[1 + \frac{\pi^2}{6} \left(\frac{a}{R} \right)^2 m(m+5) + \dots \right], \quad m = 2, 4. \quad (14)$$

Here $R = r_s \mathcal{N}^{1/3}$ is the radius of the cluster with \mathcal{N} atoms, with r_s being the Wigner-Seitz radius, which for the Na clusters has the value of 3.93 a.u.. a is a parameter defining the thickness of the diffusion region.

In this way the sum $S(\mathcal{N})$ can be written in the following way

$$S(\mathcal{N}) = \frac{9\hbar^2 e^2}{2m} \left[\mathcal{N} - \frac{6}{5} \mathcal{N}^{2/3} + \frac{99}{175} \mathcal{N}^{4/3} + \frac{\pi^2 a^2}{5r_s^2} \left(\frac{594}{35} \mathcal{N}^{2/3} - 14 \right) \right] \equiv \frac{9\hbar^2 e^2}{2m} \mathcal{F}(\mathcal{N}) \quad (15)$$

One notices that besides the number of atoms dependency, this expression involves only universal constants, which makes Eq. (15) to be, indeed, a real sum rule.

Contrary to the case of nuclear systems, where the thickness of the diffusion region is approximately the same for all nuclei, for atomic clusters the parameter a is expected to have a \mathcal{N} dependency due to the long range character of the two body interaction. This dependence was determined by interpolating the values of a satisfying Eq. (15) for $8 \leq \mathcal{N} \leq 40$. The result is:

$$a(\mathcal{N}) = -0.975157 - 0.0112138\mathcal{N}^{1/3} + 0.360518\mathcal{N}^{2/3}. \quad (16)$$

Inserting the expression (16) of $a(\mathcal{N})$ in Eq.(15), one obtains the final expression for the sum rule.

The sum rule is always a serious test for any model calculation and in particular for the RPA formalism of Ref.[10] which uses a projected spherical single particle basis, appropriate for a unified description of spherical and deformed clusters. The energy weighted sum of the dipole transition probabilities was calculated with the RPA energies and matrix elements for the transition operator. The right hand side of the sum rule equation, called $S(\mathcal{N})$ was alternatively calculated with the expressions (12) and (15), respectively. The two sets of results corresponding to the mentioned options for $S(\mathcal{N})$, are plotted in Figs. 1 and 2, respectively.

The RPA calculation of $S(\mathcal{N})$ does not yield an explicit \mathcal{N} dependence. However, this dependence is involved in the many body formalism by means of the Fermi energy, the oscillator length, the matrix elements in the projected single particle basis, the space of the single particle states used in the RPA calculation. Note that Fig. 1 shows a good agreement for medium clusters. The deviation is significant for large as well as for small clusters although both curves exhibit similar pattern concerning the oscillating behavior. The discrepancies may indicate that higher order boson expansion terms are necessary in order to improve the agreement. However, these terms would bring a certain inconsistency to the formalism since the energy weighted sum is evaluated within the RPA approach and therefore is a quadratic expression of the dipole phonon amplitudes, while higher order terms of $S(\mathcal{N})$ would be higher order polynomials of the mentioned amplitudes. Moreover, in order that Eq.(12) plays the role of a sum rule it is necessary that $S(\mathcal{N})$ exhibits a model independent expression. On the other hand adding higher order terms to $S(\mathcal{N})$ increases the specific feature of the chosen RPA approach. In contradistinction to these aspects, the second procedure described above, yields indeed, a model independent expression for $S(\mathcal{N})$. The results for this situation are shown in Fig. 2.

Fig. 2 indicates a very good agreement between $S(\mathcal{N})$ and the energy weighted sum. Actually this is the main result of our investigation. The sum rule for the modified dipole operator is equal to $S(\mathcal{N})$ given by Eq.(15) with the diffusion parameter a from (16). One notices that $S(\mathcal{N})$ is a six order polynomial in $\mathcal{N}^{1/3}$. The second remark refers to the fact that the new sum rule is different from the TRK sum rule, which is linear in \mathcal{N} .

Now we address the question whether the experimental

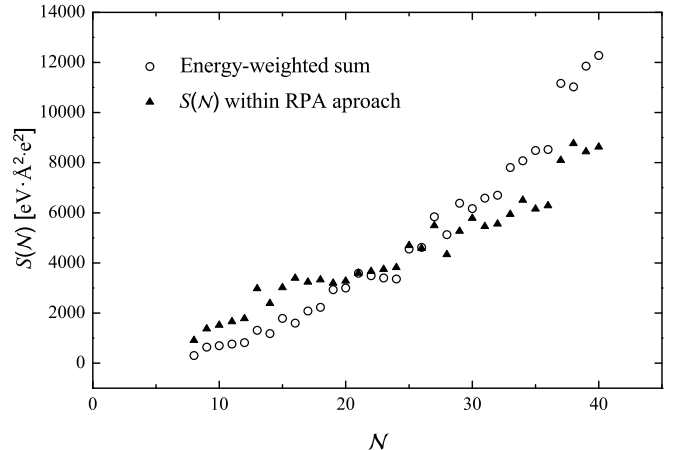


FIG. 1: The calculated energy-weighted sums of the reduced dipole transition probabilities (open circles) for Na clusters with 8-40 atoms are compared with the numerical results of $S(\mathcal{N})$ given by the RPA approach (black triangles).

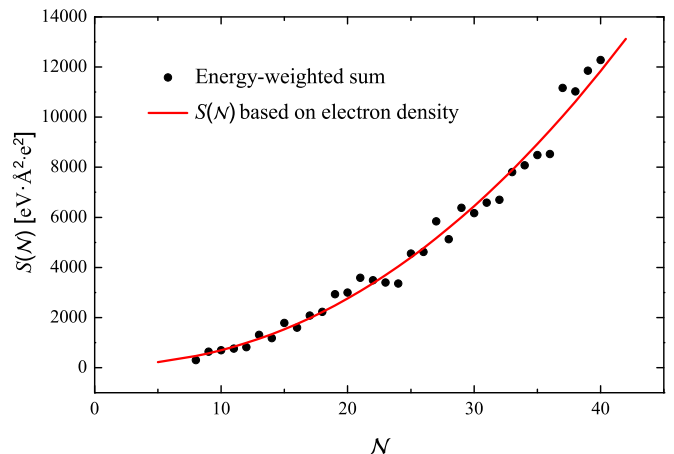


FIG. 2: The calculated energy-weighted sums of the reduced dipole transition probabilities (black circles) for Na clusters with 8-40 atoms are compared with the numerical results of $S(\mathcal{N})$ given by Eq. (15) and represented by a solid line.

data for the energy weighed sum (*EWS*) agree with the calculated values. Of course the theoretical dipole *EWS* depends on the dimension of the single particle space involved in the calculation. If the single particle states have a complex structure, as for example happens in Ref.[8], then the states with $\Delta\mathcal{N} = 3$ are connected by the standard dipole operator and consequently a large number of major shells are necessary in order to saturate the sum rule. If a projected spherical single particle basis is used,

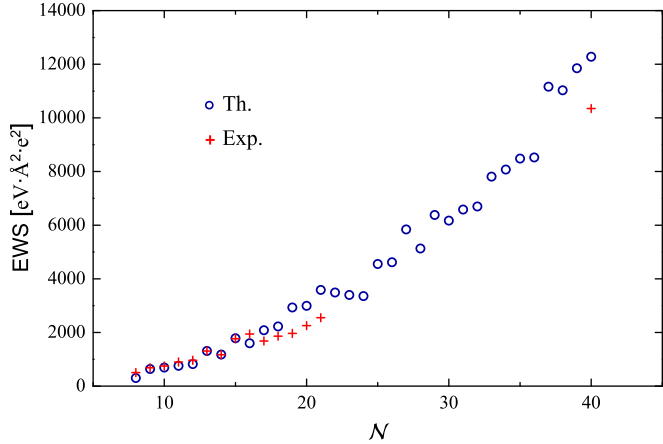


FIG. 3: The calculated (*Th.*) energy-weighted sum (*EWS*) of the Schiff-like dipole transition probabilities (open circle) for *Na* clusters are compared with the corresponding experimental data (cross) extracted from the available data concerning the photoabsorption cross section.

as we did in Ref.[10], the dipole matrix elements with $\Delta N = 3$ are vanishing. In order to account for the dipole strength brought by the non-vanishing $\Delta N = 3$ matrix elements we use an effective dipole operator which is the Schiff-like dipole operator. Therefore, the Schiff dipole moment is what we propose to describe the experimental dipole *EWS*. In order to complete the picture we have to know the experimental value for the *EWS* rule and check whether this confirms that the found sum rule is valid. Actually, this can be extracted from the experimental photoabsorption cross section. Indeed, interpolating the discrete values of the experimental photoabsorption cross sections, given as function of the excitation energy, by a smooth curve and integrating the result with respect to the energy in the interval $[0, \infty)$, one obtains the area $\mathcal{A}(\mathcal{N})$. The experimental *EWS* is proportional to $\mathcal{A}(\mathcal{N})$. We assume that the \mathcal{N} depending proportionality factor is the same as for the calculated *EWS*, i.e. $\mathcal{F}(\mathcal{N})$. This assumption is grounded by the fact that $\mathcal{F}(\mathcal{N})$ is a model independent quantity and moreover assures similar normalization for the total cross section as in the schematic calculation. Thus the experimental *EWS* is defined by:

$$[EWS]_{Exp.} = \mathcal{G}\mathcal{F}(\mathcal{N})\mathcal{A}(\mathcal{N}). \quad (17)$$

The quantity $\mathcal{A}(\mathcal{N})$, extracted from the data of Refs.[26, 27], varies between $0.582 \text{ (eV)}\text{\AA}^2$ for $\mathcal{N} = 9$ and $0.387 \text{ (eV)}\text{\AA}^2$, for $\mathcal{N} = 19$. Here the constant factor is $\mathcal{G} = 70.1591912[e^2]$. This value was obtained by equating the two expressions of *EWS*, for one chosen cluster. Thus, the constant \mathcal{G} yields a normalization of *EWS* which accounts for the "missing sum rule", noticed experimentally [28]. In Fig.3 this quantity is compared with the theoret-

ical weighted sum calculated within the RPA approach, which uses a projected spherical single particle basis [10],

$$[EWS]_{Th.} = \left[\sum_n E_n B(0^+ \rightarrow 1_n^+) \right]_{RPA} \quad (18)$$

The agreement between the two *EWS*'s, shown in Fig. 3, is a guarantee that $[EWS]_{Exp.}$ obeys, indeed, the equation:

$$[EWS]_{Exp.} = \mathcal{S}(\mathcal{N}). \quad (19)$$

Finally, we want to mention that the octupole correction to the dipole transition operator was also used in connection with the description of the electric dipole transitions in nuclear systems. Thus, in Ref.[21] it is pointed out that adding the octupole correction, the agreement with experimental data concerning the E1 transitions is substantially improved. In Ref.[22] a similar effect is obtained by modifying the many body wave function due to the octupole interaction which is considered in addition to an isovector-dipole interaction. The new components are connected by the standard dipole operator and consequently modifies the E1 transition rates. Of course, an energy-weighted sum rule associated to the dipole transition operator holds also for nuclear systems. Obviously, changing the transition operator, as it happened in Ref.[21], the corresponding sum rule which should be valid is the one obtained in the present letter.

Another interesting example is the $N - Z$ sum rule, which holds for the single beta transition. This rule says that for a single beta decaying nucleus, the difference between the β^- and β^+ strengths should be equal to $3(N - Z)$. This sum rule is true, for example, for Gamow-Teller (GT) dipole transitions and is exactly satisfied within the proton-neutron quasiparticle RPA approach. Extensions of the microscopic formalisms to the double beta decay $2\nu\beta\beta$, showed that in order to describe the transition rates, it is necessary to improve the wave functions of the mother nucleus as well as the GT dipole states, by adding anharmonic effects. However, these corrections violate drastically the $N - Z$ sum rule. In the spirit of the present paper, we open the question whether the Gamow-Teller proton-neutron interaction could be extended by adding an octupole component such that to the new proton-neutron interaction a modified $N - Z$ sum rule corresponds. This would make the inclusion of anharmonic effects which, as a matter of fact, violates the Pauli principle, unnecessary.

The final conclusion is that the Schiff-like dipole moment used for the RPA description of the photoabsorption cross section spectrum, satisfies an extended TRK dipole sum rule. The saturation of the extended sum rule is a positive test for the single particle basis as well as for the dimension of the dipole *ph* space involved in the RPA description. Also we showed that the experimental value of *EWS* is reproduced to a high accuracy by the sum rule $\mathcal{S}(\mathcal{N})$. We pointed out that the extended TRK sum rule is of a general interest, being applicable also for other

many body systems correlated by a Schiff-like two body interaction. The fact that sum rules are of paramount importance in exploring the many body properties mirrored by the multipole electric, or magnetic, transitions have been stressed by many authors [23, 24, 25]. Here we showed that a sum rule may hold also for a multipole mixed transition operator.

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